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REVIEW OF SELECTED ARMY-FUNDED RESEARCH ON FOG OIL SMOKE CHARACTERISTICS AS RELATED TO CLEAN AIR ACT ISSUES

Nancy A. Chester

RESEARCH AND TECHNOLOGY DIRECTORATE

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The endurance of fog oil (FO) as a choice training smoke is credited to its advantageous characteristics, which lead to optimum obscuration of troops and mechanized equipment as well as its cost efficiency, safety, and ease of handling. Fog oil's popularity, however, has been impacted by environmental regulation of smoke aerosol emissions under the Clear Air Act (CAA). Potential violation of the CAA by use of FO has lead to a request for further understanding of technical data describing the characteristics and behavior of FO droplets/particles and plumes. Selected sources of Army-funded research have been reviewed and summarized in this report. The impact of FO aerosols on the CAA is described according to categorized variables, which together, determine the dissemination and dispersion/disposition characteristics of FO aerosols. The variables are categorized by (1) source (smoke release point variables), (2) meteorology, and (3) plume (concentration distribution). The fate of fog oil is reported in relation to its deposition and persistence.

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EXECUTIVE SUMMARY

The ability of FO smoke disseminations to negatively impact CAA regulations is found to depend on the variables which characterize dispersion and deposition of the smoke cloud and which may be categorized by source, meteorology and plume. A summary of some of these variables and other subjects is presented in Table 4 of the full report. Source variables such as particle-size distribution contribute to the persistence of the aerosol droplets (99:1 ratio of liquid droplets to vapor) in the air by particle size influence on settling/deposition velocities, accounting for the ability of FO droplets to respond to the smallest motions in the atmosphere, acting as a passive tracer. FO aerosol droplets have been measured well inside of 10 μm with a majority of the cumulative mass at or below 2.5 μm, thereby demonstrating the potential to affect the limits of the particulate matter regulations. Fog oil aerosol's composition of known components which meet the VOC definition, and the theoretical calculation of a 30-40% decrease of mass of a single FO particle of 1.5 μm at between 0 and 40°C within an hour, suggest potential contribution of VOCs while FO is airborne, but remains to be demonstrated empirically. Although it has been shown that FO components revolatilize from surfaces should deposition occur, significant contributions to ambient VOC concentrations from revolatilization during field disseminations are questionable due to FO's particle size distribution which results in low deposition and settling velocities, as well as the element of absorption of the FO components into foliar surfaces following any deposition.

Meteorological conditions such as dispersion and micrometeorology influence the behavior of a FO plume and will affect CAA compliance within and outside of installation boundaries based on aerosol concentration and deposition level predictions (plume) from aerosol models. The complexity of meteorological conditions and their influence are represented by dispersion variables such as mean wind, turbulent diffusion, convection, and microscale meteorological variables constituting surface terrain, site geography and temperature gradients. These factors have been empirically demonstrated to readily affect FO behavior and travel, with direct influence on ground-level FO aerosol concentrations. Alteration of local microscale climate by use of several generators leading to a smoke plume capable of reducing thermal convection by way of blocking the solar heat flux, has been theorized to potentially increase ground-level aerosol concentrations. Predictive models lacking the ability to accept and incorporate a broad list of influencing meteorological variables which realistically represent training/use conditions, have been shown to lack the parameters to generate accurate predictions.

Models may be validated by comparison of predictions to field data generated under the same or similar conditions as represented by model inputs, in order to investigate their accuracy and limitations. These field data include aerosol concentrations and deposition rate/levels at positions of smoke-cloud impact, and are variables constituting smoke plume definition which will impact CAA standards based on air concentrations, and the removal of FO from the atmosphere by deposition. In general, estimated aerosol concentrations in appear higher at comparable stability classes and distances downwind from the generator, than empirical data, yet

the comparison is complicated by differing flow rates, numbers of generators used and duration times. Concentration-reducing factors are reported to have been neglected by the models used. A need for concerted and prioritized efforts toward investigation and use of validated and most representative models, in order to provide end users with the most accurate estimations possible, is readily apparent.

Once disseminated, the fate of FO is dictated by source and meteorological variables, and FO's physical and chemical properties, leading to plume travel which may be far downwind from the source under stable conditions, and/or dispersion of the aerosol particles until they can no longer be measured or meteorological conditions dictate their removal, perhaps by impaction from increased wind speeds. Deposition is reported to be dependent, in part, on particle size where sizes outside of 0.1 - 1.0 μm are more readily deposited. Negligible deposition to samplers beyond 25 meters in field studies where the MMD of FO was 1.0 μm, and measurable deposition to environmental and surrogate receptor surfaces in a wind tunnel setting with particles at an average MMAD of 2.33 μm, are examples of FO data that support dependence on particle size for deposition. Higher calculated deposition velocities (V_d) are attributed to larger particle sizes of aerosols. As entries into dissemination models that predict deposition levels, V_ds must accurately represent the particle-size distribution found during actual training and testing for a model to provide realistic estimates, and not those which are, for example, based on FO particle-size distributions which are the result of coagulation from use of unrealistically high concentrations or those measured prior to thermal equilibrium during dissemination.

In cases where FO may be deposited during times of use, residues are not expected to persist. Volatility of FO from surrogate and environmental receptors has been reported, and reduction in mass of FO aerosols has been theoretically calculated, although not measured. FO introduced into terrestrial and aquatic environments either as aerosol residues or as neat liquid, is expected to lessen in time by undergoing several modes of biodegradation and will be dependent upon a variety of factors which will determine the length of time for complete degradation.

PREFACE

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CONTENTS

TABLES

REVIEW OF SELECTED ARMY-FUNDED RESEARCH ON FOG OIL SMOKE CHARACTERISTICS AS RELATED TO CLEAN AIR ACT ISSUES

1. INTRODUCTION

Use of fog oil (FO) as a smoke screen to mask movements of troops and mechanized equipment has been employed since World War II and continues to be heavily used due to its low cost, optimum effectiveness as an electromagnetic obscurant in the visible spectrum, dispersion characteristics, ease of handling, and safety (Driver et al., 1993). Military specifications for FO continue to be updated (from MIL-SPEC "A" to MIL-SPEC "E") to provide the safest form of FO obscurant to man and environment. Furthermore, mechanical smoke-generator systems continue to be modified and improved to provide troops with products meeting modern military requirements; the use of FO with other materials, such as graphite flakes, has been found to extend the effectiveness of the generator emissions to the infrared spectrum.

Although the many advantageous characteristics of FO aerosols alone or in combination with other materials account for its endurance as a choice training smoke, the necessity of the armed forces to continue with its use in order to maintain the readiness of its troops has been impacted by Environmental Regulators' increasing concerns over potential violations of the Clean Air Act (CAA) from emissions of smoke aerosol, including those of fog oil. Training with FO has been restricted or halted in situations where opacity, particulate matter, hazardous air pollutant or volatile organic compound standards of the CAA were deemed in potential violation. In order to address Regulators' issues, and provide environmental and program managers with a more thorough understanding of FO smoke behavior, a review and interpretation of a selection of the Army's technical research data on FO aerosol with respect to the CAA, is presented in this report.

Determination of impact of FO aerosols begins with consideration of its dissemination and dispersion/deposition characteristics. These characteristics are determined by many variables, many which have been previously defined in order to describe a minimum set of measurements needed to produce an ideal data set for use in evaluation of aerosol dispersion models (Policastro and Dunn, 1985). Policastro and Dunn (1985) categorized the variables by 1) *source* (location, elevation, and exit diameters of the smoke release points, direction of smoke plume release, mass release rate, duration of operation, exit temperature, velocity of smoke plume as a function of time, particle size distribution and composition of material as it leaves generator, angle of exhaust flow, and initial effective plume height), 2) *meteorology* (wind speed and direction, stability class, roughness height (affects of buildings/vegetation), friction velocity (measure used to define meteorology of the boundary layer), mixing height, vertical variation of temperature with height) and 3) *plume* (concentration distribution). These categorizations provide an outline with which to summarize and better understand technical data from selected Army-funded research articles on FO-aerosol behavior and fate with respect to the Clean Air Act.

2. SOURCE

The generation systems designed for dissemination of fog oil, and their unique operation, directly affect variables defined by source (Driver et al., 1993; Policastro and Dunn, 1985), thereby contributing to dispersion characteristics of a fog oil aerosol cloud. Basically, all smoke generation systems used for producing fog oil plumes have provided for the vaporization of the liquid oil in hot exhaust or combustion gases, and its subsequent recondensation once exited via nozzles or exhaust pipes into the cooler atmosphere, resulting in the formation of micron-sized oil droplets. Pulse-jet engines of the M3A4 (gasoline-fired) (Liljegren et al., 1989), M3A3E3 (diesel-fired) (Liljegren et al., 1988; Policastro et al., 1989a), and M3A3 (Brubaker et al., 1992)) generators, and gas turbine engines of the XM56 generator (Driver et al., 1991; Driver et al., 1993) and SG18-02 (Nester and Imsand, 1993), are among those generators previously used to create visual obscuration by disseminating white fog oil clouds.

Most recently, the army has begun to field the M56, a dual-purpose motorized smoke generator for large area visual and infrared smoke obscurations, and the M58, a mechanized smoke generator for visible and infrared screening. The M56 and M58 share the same multispectral smoke generator technology that is powered by a turbine engine and which provides both hot exhaust for vaporizing fog oil, and bleed air that powers an air ejector for disseminating infrared obscurants. The system can provide visual, infrared or combined visual/infrared screening at variable rates. Variation in operation of FO-generating systems, including the M56 and M58, will control and/or influence several source variables (Driver et al, 1993; Liljegren et al, 1988; Liljegren et al., 1989) including tactical source variables such as location, elevation, and direction of release, as well as angle of exhaust flow, duration of operation, and initial effective plume height (as determined from exit temperature and velocity of plume as a function of time (Liljegren et al., 1989; Policastro and Dunn, 1985)). Operational variables affecting source definition include the mass release rate (maximum: 1.33 gal/min), exit temperature, composition of FO (particle:vapor ratio) as it leaves the generator, and particle-size distribution. Source variables under consideration for compliance with the Clean Air Act include aerosol particle-size distribution in relation to particulate matter (PM-10, PM-2.5) standards, and composition of material as it leaves the generator in relation to contribution to volatile organic compound (VOC) standards.

2.1 Particle-Size Distribution

Particle-size distribution (PSD) refers to the frequency of particle occurrence as a function of particle diameter and particle occurrence may be expressed as mass or particle number (Cataldo et al., 1989; Driver et al., 1993). Fog oil aerosols are identified as having a log normal distribution of particle sizes (Liljegren et al., 1989) that, like other aerosol PSDs, are affected by exit temperature from the generator and the concentration of the aerosol and so may vary accordingly. The exit temperature of the vaporized FO, and therefore the dynamics of condensation, are affected by both the generation method (Driver et al., 1993) which influences the temperature at which liquid FO is vaporized, as well as the flow rate of the oil where lower

rates of release increase exit temperatures, while higher flow rates decrease temperatures (DeVaull et al., 1989). When exit temperature is increased, the mean particle diameter is shown to be reduced (DeVaull et al., 1989, Liljegren et al., 1989). Aerosol concentration influences the rate at which FO droplets coagulate (Driver et al., 1993) where higher concentrations increase coagulation, thus increasing particle size (Cataldo et al., 1989). Particle size influences mechanisms of deposition (Liljegren et al., 1989) and is among the properties of FO that determines its persistence in the atmosphere (fog oil fate is further discussed in Section 4.2). Log normal PSD may be characterized by reporting two parameters: 1) a mean or median size such the mass median diameter (MMD) or mass median aerodynamic diameter (MMAD) and 2) the geometric standard deviation. For log normal distributions, the median diameter equals the geometric mean diameter for both mass and particle number (count) frequencies (Reist, 1993).

Particle-size distribution of FO aerosols has been measured by several authors. Studies of PSDs from outdoor field studies conducted by researchers at Argonne National Laboratory and the University of Illinois at Urbana-Champaign, at three different sites (Liljegren et al, 1988; Liljegren et al, 1989; Maloney et al, 1992), measured MMADs ranging from 0.8 to 1.0 μm (Policastro et al, 1989a) with geometric standard deviations from 1.5 to 1.7. Based on log-probability graphs of distribution of mass as a function of diameter, and histograms of probability densities, percent of the cumulative mass less than or equal to diameters of interest may be approximated. For field trials at the Atterbury Reserve Forces Training Center test site near Columbus, Indiana, approximately 98% of FO particles by mass, occurred between 0.3 and 3.0 μm (Liljegren et al., 1989). Graphs of PSD plotted for field tests conducted at Dugway Proving Ground in Utah were similar with approximately 92% of the cumulative mass ≤ 4.5 μm and 83% ≤ 2.5 μm (Liljegren et al., 1988). A companion paper to the field-test reports which describes analysis methodologies, measured particle size distribution in a laboratory setting for comparison to field data, and measured the MMAD at 0.8 μm and reports 99% of FO particles ranging from 0.1 - 4.0 μm (DeVaull et al., 1989).

Data generated from field studies during aerosol product trials for the characterization of smokes as well as evaluation for use in models for smoke hazard assessment, are reported as both particle-size distributions in number (count) fractions, as well as MMDs (Policastro and Dunn, 1985). One hundred percent of the particles were determined to be under 5.0 μm and at least 96% were ≤ 3.0 μm. High MMDs are reported at 3.9 μm and 4.9 μm during use of the XM49 and M3A3 generators, respectively, and do not compare well with aforementioned field data. Data from the Smoke Week III trials are considered questionable (Policastro and Dunn, 1985) due to problems with quality and accuracy of sampling, as well as a revealed evolution of particle-size distribution over the time it took the M3A3 to reach thermal equilibrium. Sampling of particle size prior to achievement of thermal stabilization is considered the likely cause of the reported high MMDs, and agrees with affects of temperature on particle size (DeVaull et al., 1989; Liljegren et al., 1989).

Studies conducted in a wind tunnel at the Pacific Northwest Laboratory by Cataldo et al. (1989) measured MMADs (GSD) ranging from 1.6 μm (1.7) to 3.1 μm (1.7); the authors

consider the higher PSD measurements to be the result of the influence of increased aerosol mass concentration (up to 990 mg/m^3), which resulted in increased particle sizes due to coagulation of aged aerosols in the wind tunnel. Graphs of particle-size distributions indicate no measurements of particle sizes 10 μm or above and extrapolation of the data indicates an approximate average of 98% of the cumulative FO mass ≤ 10 μm and 30-85% < 2.5 μm (lower percentages indicative of the higher MMADs). Cataldo et al. (1989) report that their PSD measurements fit within a range of previously measured MMADs of 0.6 - 3.0 μm, although their data are at the upper extreme.

Previously measured particle-size distributions are reported throughout the cited literature, and are the result of differing dissemination conditions, measurement techniques and instruments. A literature review by Brubaker et al. (1992) documents reported measurements of average mass mean diameters ranging from 0.74 to 1.68 μm; Cataldo et al. (1989) compare their wind tunnel data to a previously reported MMAD range of 0.6 to 3.0 μm and Liljegren et al. (1988) compare their field data to mass mean and mass median diameters of 1.16 and 0.9 μm, respectively. In the literature review by Driver et al. (1993), the authors refer to 1) previously measured MMADs (GSD) from 0.6 (1.6) to 1.3 μm (1.6), and 2) MMDs from 0.7 to 1.7 μm, as well as 3) count median diameters of between 0.5 and 1.0 μm. The authors used these data in order to arrive at a "typical" aerodynamic size distribution of FO aerosol characterized by a MMAD of between 0.9 and 1.9 μm, with 1.4 μm and a GSD of 1.65 as the representative values which were subsequently employed for calculation of coagulation, dispersion, and deposition. Clearly, the particle size of FO aerosol droplets will be a CAA regulatory concern for as long as they remain in the air.

2.2 Volatile Organic Compounds (VOCs) in Fog Oil

The definition of a VOC is any organic compound of carbon (excluding carbon monoxide, carbon dioxide, carbonic acid, metallic carbides or carbonates, and ammonium carbonate) which participates in atmospheric photochemical reactions. A list of further exclusions due to negligible atmospheric photochemical reactivity, is listed in 40 CFR Ch. 1, Part 51 (*Code of Federal Regulations*, 1996) under the definition of a VOC. The components that are known to make up FO fall within this definition both as compounds of the liquid which reportedly undergoes negligible gross changes (DeVaull et al., 1989; Nester and Imsand, 1993) during transition to aerosol material, and also as compounds forming the resulting aerosol which consists of a two phase mixture of micron sized liquid droplets and vapor at a ratio of 99:1 (DeVaull et al., 1989). Analyses conducted (Brubaker et al., 1992; Cataldo et al., 1989; DeVaull et al., 1989, Driver et al., 1993; Nester and Imsand, 1993) determined FO liquid and aerosol to consist of several thousand individual hydrocarbon components which meet the VOC definition, the majority of which are present in quantities less than 0.1%, including straight and branched aliphatics, as well as 2- and 3-membered ring aromatic compounds between C_{12} and C_{22} (phenanthrene, dihydrophenanthrenes, fluorenes, acenaphthene) (Brubaker et al., 1992; Driver et al., 1993).

In April of 1986 the Military Specification for FO was amended to require the removal of carcinogens, and was published as MIL-F-12070C, which has since been revised two more times.

This requirement necessitated that the oil be treated to remove carcinogenic compounds either by severe hydrotreatment, where carbon-carbon double bonds are hydrogenated and heterocyclics undergo ring opening, or by solvent refinement, in which aromatic compounds, including heterocyclics and polynuclear aromatic hydrocarbons, are removed by solvent extraction. Fog oil produced before the 1986 amended specification is referred to as "old" fog oil whereas that produced after the amendment and which has undergone removal of carcinogens, is termed "new" fog oil (Brubaker et al., 1992; Driver et al., 1993). Results of analyses of new FO would theoretically identify only aliphatic hydrocarbons since the aromatics should have been removed. The results of FO analyses in the papers reviewed for this report indicate use of old FO due to testing of samples found to consist of 40-50% aromatics (Brubaker et al., 1992; DeVaull et al., 1989; Driver et al., 1993).

Liquid fog oil stored in vessels is considered to have the potential to contribute to VOC emissions if present in sufficient quantities (Driver et al., 1991), and the potential of FO aerosols to contribute substantially to this category of emissions either following any deposition or while still in the air, has been investigated. FO residues deposited in wind tunnel studies (Cataldo et al., 1989; Driver et al., 1991) are shown to have volatilized from surfaces of vegetation, soil, and surrogate receptors such as filters, which would provide a source of VOCs, but absorbance of FO constituents by dissolution of waxy cuticles of leaf surfaces is also suggested as contributing to measured FO losses during persistence studies (Driver et al., 1991). Revolatilization of FO residues following deposition is also reported by Dunn et al. (1996), and a study of continued aspiration of absorbent tubes during sampling of FO-aerosol's vapor fraction resulted in measurable evaporation (DeVaull et al., 1989). Nester and Imsand (1993) report that FO aerosol disperses and evaporates, as opposed to condensing on the ground, but provide neither details of this proposed process, nor cite references from which the conclusion was based.

Calculations based on a theoretical evaporation model used by Driver et al. (1993) indicate that a FO particle of 1.5 μm, exposed to air may result in 30-40% decrease of mass within a 1-hr period at temperatures between 0 and 40°C, but were not given consideration in a dispersion modeling effort due to variation in temperatures at which FO smoke may be disseminated and the relatively short period of time the aerosol was considered to remain in the air (Driver et al., 1993). Empirical data representative of FO's particle-size distribution and effects on particles within a cloud, are not available to validate these theoretical calculations. Whether these findings indicate potential significant contributions to VOCs for regulatory purposes remains to be determined by the specifics of a proposed testing scenario and by addressing the questionable likelihood of actual deposition to surfaces of FO particles, as well as impact of any potential revolatilization once the "old" oil undergoes further refinement for removal of carcinogens thereby eliminating aromatics. Discussion of the fate of aerosol particles after dispersal is found under Section 4.2.

3. METEOROLOGY

Atmospheric conditions are known to affect FO-plume travel (Driver et al., 1991; Driver et al., 1993) and have been well-described and measured by investigators from Argonne National Laboratory and University of Illinois, Urbana-Champaign (Liljegren et al., 1988; Liljegren et al., 1989; Maloney et al., 1992; Policastro et al., 1989a) and constitute a number of descriptive parameters entered into FO-smoke dispersion models (Brown et al., 1993; Driver et al., 1991; Driver et al., 1993; Nester and Imsand, 1993) for estimation of aerosol concentrations and deposition levels downwind from stationary sources. This information is critical in determining CAA compliance within and outside of installation boundaries. Meteorological conditions affecting plume travel include dispersion and those resulting from the microscale meteorology (micrometeorology) as influenced by the surface terrain and site geography.

3.1 Dispersion

In a series of field disseminations of fog oil under varying stability classes (Liljegren et al., 1988; Liljegren et al., 1989; Maloney et al., 1992), dispersion of FO plumes from stationary sources was attributed to mean wind (speed and direction of wind), which transported the plume, and turbulent diffusion which contributed to the spread of the plume. Effects of relative (comparative) wind speed were reported. Tests with higher winds caused FO plumes to be less wide than during lower wind speeds (Liljegren et al., 1988; Liljegren et al., 1989); greater lateral wind fluctuations produced a greater lateral spreading. Lower wind speeds resulted in proportionally higher FO aerosol maximum concentrations at given downwind distances, but also were subject to effects of convection and upward momentum of plumes being augmented by strong insolations (Liljegren et al., 1988).

Analyses of data for source, meteorological, and plume measurements indicated the dominant roles in dispersion to be gradients formed from 1) mean and fluctuating wind speed components (wind vector magnitude, azimuth, inclination) with increases in height above the ground, and 2) convection caused by solar heating during the day (insolation) with radiative cooling at night. Under convective meteorological conditions, a rise or lift-off occurred downwind of the source at the centerline of the plume with the more convective conditions resulting in lift-offs closer to the generator. The effects of convection corresponded to decreases in ground-level concentrations in the field (Liljegren et al., 1989; Maloney et al., 1992) and are discussed with respect to micrometeorology in the following section.

3.2 Micrometeorology

Three categorizations of meteorological events are described in Liljegren et al. (1989) of which microscale meteorological events are one. Micrometeorological events are characterized as having horizontal distances (scales) of less than 2 km and a time scale in the range of minutes or seconds, and are strongly affected by *local* terrain, vegetation, and temperature gradients. The other categorized atmospheric phenomena include macroscale events, which arise due to global

temperature gradients between the tropics and polar regions, and include processes such as the jet stream, and mesoscale events, such as thunderstorms, which arise from heat released during cloud formation and from regional temperature gradients formed from variations in surface heating between, for example, large bodies of water and coastal regions, and/or mountain ranges and valleys.

Effects of the micrometeorology at field sites where FO was disseminated at Dugway Proving Ground (DPG) (Liljegren et al., 1988), Camp Atterbury (Liljegren et al, 1989), and Meadowbrook (Maloney et al., 1992), were apparent upon authors' data analyses. At DPG, terrain and meteorology were each categorized as simple. The terrain was homogeneously flat with no downwind changes. Fog oil plume behavior was subject to transitional and unfavorable convective meteorological conditions in late-morning and early-afternoon test periods and resulted in uncharacteristically low exposure levels at the height of release. Use of more favorable and more neutral "dawn" conditions resulted in a smoke plume confined to a narrow corridor.

Micrometeorology became more sophisticated at Camp Atterbury and Meadowbrook field sites. As was the case at DPG, terrain was categorized as simple at Camp Atterbury, but meteorology differed and was reported as complex. Rises in plume heights were reported for three FO disseminations at the Camp, but were the result of differing causes. During two runs with "highly variable" wind speed and "fairly constant" wind direction (actual parameters are available in the report), the FO plume was noted to have moved in pulses versus as a uniform cloud. Downwind from the source, the *non-uniform upwind terrain* (terrain rose up to 50 m above elevation of the site and was densely wooded) influenced meteorology by producing large-scale, low-frequency motions similar to thermal convection, which effectively produced a rise in plume height from the ground (convective lift-off). In a third run, during which time wind direction was "highly variable" and wind speed was much lower than previously, a plume lift-off from the ground occurred closer to the release point as a result of *thermal* convection. Incidence of thermal convection was supported by values determined from calculations that described a planetary boundary layer supportive of thermal convective behavior for the third test while only some convective influence would be anticipated for the other two tests (Liljegren et al., 1989).

The authors go on to note that the local microscale climate may potentially be altered during strongly convective conditions, wherein the combined smoke plume from use of several generators can shield the ground from the solar heat flux. Less thermal convection could effectively lead to greater ground-level concentrations than would be expected from use of a single generator where the concentration field is at a higher, superposition (Liljegren et al., 1989).

The Meadowbrook site was characterized as having both complex terrain and complex meteorology. The site was located in the foothills of a mountain chain and consisted of a forked creek valley with rising surrounding slopes. Micrometeorology was characterized by a density-driven, diurnal upslope-downslope flow pattern of air during extreme temperatures of day and night, respectively (Maloney et al., 1992). The site was chosen to examine FO smoke releases under stable, nighttime/early morning conditions of downslope wind conditions, although both

diurnal extremes were measured. Results of releases indicate that smoke dispersion was very sensitive to the local stability and surface roughness. Under stable conditions, the constraining effects of the valley wall limited both plume trajectory and horizontal plume growth. Much higher concentrations were produced downwind at Meadowbrook, with comparable source characteristics, than at DPG or Atterbury with only a 50 percent reduction in average concentrations over distances 200 - 1000 m. Under unstable conditions, FO-smoke dispersion was rapid; plume centerlines rose off the ground immediately at the generator to 250 m away from the source due to heat flux from the warm ground (Policastro et al., 1989a).

3.3 Other Meteorological Conditions

In a study by Cataldo et al. (1989), the effects of varied environmental conditions on ecological effects of FO aerosols on several plant species was conducted in the controlled environment of a wind tunnel. Authors were specifically interested in the correlation of test responses with dose/mass loading to receptor surfaces (plants) rather than airborne smoke concentrations, and investigated effects of relative humidity, rainout and also wind speed, during exposures at a constant temperature range of 20-23°C.

Aerosol concentrations of FO were not affected by water vapor present as high humidity in the wind tunnel atmosphere but particle size of the aerosols increased strongly as a function of increased aerosol mass concentration, attributed to the coagulation of small particles into larger ones. The larger mean particle size is reported to account for the significant deposition velocities and subsequent mass loads observed in this study. Deposition is discussed in section 4.2.1 below, and may not occur as described by Cataldo et al. (1989) during field disseminations where coagulation may not readily take place and mean particle size has been reportedly smaller.

Under the testing conditions of a simulated rainfall, results indicated no significant reduction in mass load when compared to a control; no displacement of FO from plant surfaces was shown, and deposition continued in the presence of the simulated rainfall. Wind speed was tested with respect to its affect on FO-deposition, and results are discussed in the section on deposition. Due to the wind tunnel environment, no other factors contributing to the dispersion of a FO cloud were tested for potential effects..

3.4 Dispersion Models

Affects of atmospheric conditions and terrain are considered by dispersion models when meteorological variables, such as wind speed and direction, stability class, roughness height (dispersion coefficients taking into consideration affects of buildings and vegetation), friction velocity (measure used to define meteorology of the boundary layer), mixing height and vertical variation of temperature with height (Driver et al., 1991; Driver et al., 1993; Nester and Imsand, 1993; Policastro and Dunn, 1985; Policastro et al., 1989b) are entered into the dispersion equation. Less sophisticated models, such as generic Gaussian-plume dissemination models, may not be designed to accept such an extensive list of variables as input, and have been shown to lack

the ability to predict convective plume lift-off, and subsequently overpredict both the maximum FO-aerosol concentration and plume width at ground level (Liljegren et al., 1989). Further discussion of issues concerning dispersion models is presented in the section 4.1.

4. PLUME

The variables constituting smoke plume definition include aerosol concentration and deposition rate/levels at positions of plume impact, as well as particle size distribution (Policastro and Dunn, 1985). It is upon the determination of the former two particular values that many ultimate decisions regarding training and use of FO are based, with respect to potential adverse effects to both man and environment, and which have been measured in field studies and repeatedly estimated by use of a variety of dispersion models.

4.1 Fog Oil Aerosol Concentrations

Fog oil aerosol concentrations downwind from generators are of prime importance due to the potential impact of disseminated obscurants on Clean Air Act standards which regulate emissions based on air concentrations, per time-averaged basis, of components of interest such as particulate matter 10 and 2.5 μm or below, or opacity - the degree to which emissions reduce the transmission of light and obscure the view of an object in the background. All of these standards incorporate the concentration of the obscurant, thus both field and wind tunnel concentration measurements are presented, as well as estimated concentrations generated from dispersion models.

Empirical field data generated from three thorough investigations of FO dissemination and dispersion are documented in a series of reports describing studies conducted at Dugway Proving Ground, Utah; Atterbury Reserve Forces Training Center, Indiana; and Meadowbrook, California (Liljegren et al., 1988; Liljegren et al., 1989; Maloney et al., 1992). Other documented field studies have been conducted at Eglin Air Force Base as part of Smoke Week III and IV, for the purpose of aerosol product trials, but which produced data of questionable integrity due to a host of methodology problems, and so will not be considered in this report (Policastro and Dunn, 1985). Further successful efforts to provide quality data are presented in the companion document for the field studies where oil analysis, including that from aerosol samples, is described and tailored to the authors' research needs (DeVaull et al., 1989).

Concentration data for the field studies at Dugway Proving Ground and Camp Atterbury were collected under near-neutral to very unstable atmospheric conditions using one M3A3E3 and one M3A4 generator, respectively. Collection of aerosol samples at Meadowbrook took place during extremes of atmospheric stability - unstable and stable conditions only, using one M3A4 generator. For all sites, generators were stationary, duration of FO disseminations ranged from 10 minutes to 1 hr, and average flow rates ranged from 0.523 - 0.762 gal/min. Samples were taken at various distances downwind from the generator (source) by the use of aspirated Tenax- (chemical adsorbent which removes hydrocarbons from the air flow drawn through the tube)

filled samples tubes (DPG) and cassettes containing 37 mm glass fiber filters (Atterbury and Meadowbrook). Sampling procedures were modified and improved following results from the earliest (DPG) investigation. Analysis was performed by high temperature thermal desorption procedures followed by gas chromatography for chemical characterization and quantification (DeVaull et al., 1989; Liljegren et al., 1988; Liljegren et al., 1989; Maloney et al., 1992).

Concentrations presented in Table 1 are time-integrated (smoke concentration is averaged over the total duration of the test) averages of concentrations from three or more trials per site. Concentrations in mg/m^3 ranged a) from 1.6-120 at 25 m to 0.05-3.6 at 200 m at DPG, b) from 0.024-76 at 50 m to 0.016-0.13 at 675 m at Atterbury, and c) from 0.44-100 at 25 m to 0.015-1.2 at 250 m under unstable conditions at Meadowbrook with from 0.065-36 at 25 m to 0.011-21 at 250 m to 0.023-0.21 at 4000 m during stable conditions. Instantaneous plume concentrations (those not averaged over the duration of the test but taken instantaneously) include periods of higher smoke concentrations at as much as a factor of 10 or more (Dunn et al., 1996; Policastro et al, 1989a) as well as smoke-free periods not apparent when averages are reported.

TABLE 1. Empirical Aerosol Concentrations of Fog Oil

FIELD DATA
(mg/m^3)

KILOMETERS

	.025	.050	.10	.13	. 20	.25	.45	.60	.68	2.0	4.0
DUGWAY near-neutral to very unstable average = 1 hr duration average = 0.523 gal/min	1.6-120	0.68-30	0-7.7		0.05-3.6						
CAMP ATTERBURY near-neutral to very unstable average = 52" duration average = 0.628 gal/min		0.024-76	0.036-24			0.030-3.0	0.025-0.88		0.016-0.13		
MEADOWBROOK duration: 10-45" per trial Unstable	0.44-100			0.019-2.6		0.015-1.2					
Stable average = 0.762 gal/min	0.065-36			0.017-16		0.011-21		0.093-11		0.072-2.5	0.023-0.21

In contrast to field studies of FO, a study by Cataldo et al. (1989) describes FO aerosol production using a laboratory generator which dispersed the smoke into a wind tunnel (operated as a continuous loop) for use in studies designed to investigate the mechanisms controlling

transport, fate, and ecological effects of FO under a variety of controlled environmental conditions. The combination of the design of the generator and wind tunnel allowed a continuous introduction of FO into the tunnel where the aerosol was allowed to age during exposure tests. The authors report that unrealistic aging was prevented by introduction of a flow of carrier air into the wind tunnel while an equivalent flow was drawn out resulting in a mixture of freshly generated and aged particles. This design is considered by Cataldo et al. (1989) as having provided an accurate simulation of actual field conditions (average age of the aerosol in the tunnel was estimated at 41 minutes) where a cloud would be transported about 2 km downwind in a 0.9 m/s (2 mph) wind.

Fog oil aerosol was collected in the wind tunnel using aspirated 25-mm glass fiber filters for collection and a continuous record of concentration (as a relative measure of aerosol mass) was taken by means of a laser transmissometer. Samples were extracted with isooctane and analyzed via high performance liquid chromatograph (HPLC). Concentrations within the wind tunnel ranged from 700 - 1000 mg/m^3 outside of tests using cumulative dosing (Cataldo et al., 1989).

Several reviews and assessments of FO obscurants relied on smoke dispersion models to predict concentrations of aerosol clouds downwind from a source under a variety of atmospheric and generation conditions. Results of the model runs are given in Table 2. Concentrations listed were generated by three models under specified atmospheric conditions (stability categories) for distances downwind of the generator. Where applicable, atmospheric stability classes are grouped into two major categories representing 1) the most stable of conditions considered ("moderately stable", "very stable"), and 2) neutral to unstable conditions ("neutral", "neutral-unstable"). This breakdown allows comparisons to the previously described field data which was typically gathered under various unstable (DPG, Atterbury, Meadowbrook) and stable (Meadowbrook) conditions. Values in mg/m^3 represent centerline-plume, time-averaged concentrations per distance; ranges appear when more than one atmospheric category was considered as well as when a model gave predictions for use of 6 and 12 generators (TECOM Model).

TABLE 2. Model-Predicted Aerosol Concentrations of Fog Oil

MODEL DATA (mg/m^3)

KILOMETERS

		.1	.2	.25	.4	.5	.7	.75	1	2	4	10
GAUSSIAN MODEL 1 generator; 30" duration at 1.33 gal/min	neutral-extremely unstable	27-120	7.0-34		1.8-9.0		0.59-3.1		0.29-1.6	0.08-0.52	0.02-0.18	
	moderately stable	37	140		93		42		24	7.9	2.9	
TECOM MODEL 6-12 generators; 1' duration at 2-2.2 gal/min	neutral	0.048 - 3.8		2.5-3.3		0.96-2.1		0.57-1.2	0.34-0.91	0.096 -0.31	0.024-0.072	
	very stable	0-3.5		2.5-3.6		1.1-1.2		.53-1.8	.36-1.1	0.096 -0.43	.024-0.17	
RTVS MODEL 1 generator; 30" duration at 1.32 gal/min	neutral-unstable	0-14							0.1-1			-0.001 -0.07

Atmospheric concentrations for tests planned at Yuma Proving Ground were modeled by Dugway Proving Ground's Real-Time Volume Smoke Model (RTVSM) and reported in an environmental assessment by Driver et al. (1991). The model required meteorological and source inputs, assumed no FO deposition, and generated estimates for a variety of atmospheric stability conditions out to 10 km from the generator.

In a later report by Driver et al. (1993), a basic Gaussian model was employed by which FO aerosol concentrations out to 40 km were predicted. This model also required meteorological and source inputs, included a source-depletion term (accounts for reduction in total windborne mass of a smoke plume due to deposition), and generated estimates for a variety of atmospheric stability conditions.

Three estimation methodologies are reported in an environmental assessment for Eglin Air Force Base (EAFB) by Nester and Imsand (1993), with results of the third appearing in Table 2 (TECOM Model). The first method used basic inputs such as total amount of FO expended, duration of generation, wind speed, and an estimate of the area of the aerosol cloud in a basic pyramid shape, to calculate rough estimates of aerosol concentrations at 1.48 km. Average concentrations ranged from 0.45 - 0.77 mg/m^3. This was the most unsophisticated of the procedures used for estimating aerosol concentration in what is considered to be a complex set of

circumstances, and predicted values are believed by the authors to be overestimations due to the neglect of concentration-reducing factors (listed below).

In an effort to produce more accurate data, further aerosol concentration predictions were obtained by extrapolation of estimates from the previously conducted RTVS Model run performed for Driver et al. (1991) for an environmental assessment at Yuma Proving Ground, Arizona, briefly described above. The more sophisticated calculations took into consideration the previously neglected buoyancy of hot smoke, topographic features of dissemination sites, and particle deposition (Nester and Imsand, 1993). At 5-8 knot winds, FO concentrations were (mg/m^3): 4.5-7.2 at 0.1 km, 0.7-0.2 at 1 km, 0.003-0.005 at 10 km.

In a final effort to supplement and/or improve the projected effects of the proposed field tests at EAFB, a third modeling performance that more fully represented the conditions at the site of concern, was conducted by the Meteorological Division of DPG (U.S. Test and Evaluation Command), with an unidentified model that may have been the RTVSM (named the "TECOM Model"). Different meteorological and test parameter conditions, including the input of 6 and 12 generators, were identified and results were calculated for neutral and stable conditions each and are listed in Table 2.

There have been discussion and issues raised concerning the capabilities of many FO-dissemination models to provide representative predictions of FO aerosol concentrations at distances downwind of the source. Although it is outside of the scope of this report to evaluate dispersion models, a few major factors which contribute to distinguishing the behavior, and hence aerosol concentration, of a FO cloud, have been presented by the scientists at Argonne National Labs/University of Illinois, and are a result of their evaluation/validation of model performances from comparisons with clearly defined empirical data sets (Liljegren et al., 1989; Maloney et al., 1992; Policastro et al, 1989a; Policastro et al, 1989b). In a report by Dunn et al. (1996) that synopsizes the contributing factors, the authors site 1) plume lift-off under convective conditions, 2) terrain and vegetation effects, 3) transitional meteorology, 4) short release durations (versus long or continuous stack releases), and 5) deposition and revolatilization of FO, as the critical factors that many dispersion models do not incorporate, and which often lead to overestimation of environmental impacts of FO. A new model, by the name of SMOKE, developed by this same group of researchers which addresses these factors, has been introduced, described, evaluated and is documented among the reviewed reports (Brown et al., 1993; Liljegren et al., 1989), as well as in the above Dunn et al. (1996) citation for the "Real World" Air Conference.

Nester and Imsand (1993) and Driver et al. (1991) also point out concentration-reducing factors neglected by models they used such as the buoyancy of hot smoke, topographic features of dissemination sites, and particle deposition (Driver et al., 1991; Nester and Imsand, 1993). Different conditions than those in the field were noted by Driver et al. (1993) as model assumptions that may contribute to unrealistically high predictions including, 1) input of a wind vector that is constant in direction and time versus fluctuating mean wind direction and affects

contributed by terrain, 2) gradual changes in concentration as the distance from the source increases versus minima and maxima in the field, and 3) determination of aerosol concentrations assuming no loss of FO mass by deposition or impaction onto surfaces (100% surface reflection) whereas in the field, some loss of mass may be found deposited inside 20-25 meters from the generator (Liljegren et al., 1988; Liljegren et al., 1989; Maloney et al., 1992).

Several of the reviewed papers present estimated data from a few dissemination models which were used frequently in the past to generate concentration and deposition values when applicable/comparable empirical data were not available. Due to the potential impacts of FO obscurant on CAA regulations based on aerosol concentrations, it is of major importance to provide regulators and trainers with modeling tools that have been validated with empirical data as well as evaluated for accuracy on an ongoing basis as data becomes available, and which will simulate as authentically as possible the true behavior of a FO smoke cloud in order to predict more realistic aerosol concentrations.

In summary, the large body of work by researchers at Argonne National Laboratory and University of Illinois offers well defined empirical data sets against which to evaluate/validate dispersion models, and also documents actual field concentrations of FO aerosols under several atmospheric stability classes and other defined conditions. Under more controlled conditions, wind tunnel experiments were conducted using higher FO concentrations than the highest reported time-averaged value of 120 mg/m^3 at 25 m in the field at DPG. Based on an instantaneous plume concentration that may exceed the time-averaged concentration by a factor of 10 (Policastro et al., 1989a), for example 1200 mg/m^3 versus the time-averaged value of 120 mg/m^3, values similar to those generated in the wind tunnel (700-1000 mg/m^3) may occur within the first 25 meters of the source under similar field-testing conditions but will decay with distance.

4.2 Fog Oil Fate

Aspects of the fate of FO, including the potential of the aerosol to deposit to ground, foliage, and other surfaces, thereby decreasing the aerosol mass, as well as the subsequent potential revolatilization from or absorption into these surfaces, are under consideration for affects on potential particulate matter concentration reduction and contribution to VOC emissions. The persistence of FO and/or similar oils in terrestrial and aquatic media has been investigated for toxicological impacts and is described below.

4.2.1 Deposition

The fate of FO aerosols has been investigated theoretically and empirically, and particle-size distribution is reported to influence settling and deposition of FO and the behavior of the smoke plume. Theoretical studies cited in reviewed documents predict negligible deposition for particles 0.1 - 1.0 μm due to sizes in this range too large for affects of diffusion and too small to have an appreciable settling velocity (Cataldo et al., 1989; Liljegren et al., 1988). Liljegren et al.

(1989) examined theoretical deposition of a FO aerosol, made up of 1.0 μm particles (particle size based on an average mass median diameter of approximately 1 μm determined in field trials (Policastro et al, 1989a)), to the ground surface and ground-covering vegetation in terms of filtration theory (vegetation 1 m tall = filter fibers) and five mechanisms for deposition which greatly consider particle size as a factor: interception, inertial impaction, diffusion, gravitational settling and electrostatic attraction. Based on results from calculations, effects of the five mechanisms on deposition are negligible.

Liljegren et al. (1989) also investigated FO aerosol's ability to act as a passive tracer in the atmosphere to further understand its capacity for deposition. An equation to calculate response time included FO's density and a representative particle size of 1 μm to compute relaxation time or response time of FO particles. Computations indicated a greater significance of turbulent dispersion over significance for settling and thus FO particles are expected to follow turbulent conditions, responding to the smallest motions in the atmosphere, rather than settle.

Consideration of fog oil's physical and chemical properties suggests that particles may remain aloft in the atmosphere for distances far downwind until other factors (meteorological conditions) dictate their removal and/or affect dispersion to the extent that they cannot be measured (Brubaker et al., 1992; Nester and Imsand, 1993; Policastro and Dunn, 1985). These properties include not only the aforementioned particle size of FO droplets, such that they may not readily deposit, but also FO's low vapor pressure such that 1% or less (Brubaker et al., 1992; DeVaull et al., 1989) of a FO cloud is in the gaseous state, and which accounts for its persistence and therefore desirability as a visual smoke obscurant. In order to test theoretical data, researchers conducted field studies which attempted to measure deposition.

Liljegren et al. (1988), during field disseminations of FO at Dugway Proving Ground, prepared glass fiber filters (inert), 123 mm in diameter, and mounted them on 7/16" steel posts in both vertical and horizontal orientations (for deposition and impaction) at 0.5 m in height at positions 25 to 800 m downwind of the source. Following collection, filter papers were shipped in sealed glass vials and underwent both analysis by hexane extraction/gas chromatography with a packed column, and thermal desorption/gas chromatography with both methods yielding similar results.

No significant deposition amounts for either vertically or horizontally oriented filters was found within resolution limits beyond approximately 25 meters. These results are consistent both with the mathematical determinations of insignificant deposition mechanisms for aerosol particles averaging 1 μm, and with the exceedingly small or minimal deposition proposed for particles in the range of 0.1 - 1.0 μm.

Cataldo et al. (1989) further lend evidence to indicate particle deposition *outside* of the 0.1 - 1.0 μm range with empirical data that shows FO particles larger than 1.0 μm to be strongly affected by wind speed and gravitational forces, leading to deposition. During wind tunnel studies at the Pacific Northwest Laboratories, which investigated mechanisms controlling transport, fate

and effects of FO aerosols, particle-size as MMAD (GSD) for FO ranged from 1.6 μm (1.5) to 3.1 μm (1.8) with an average MMAD of 2.33 (1.67). This average measurement is in contrast to the average mass median diameter (MMD) of 1 μm in previously described field studies.

Cataldo et al. (1989) *did* find FO deposition to plant, soil and surrogate receptor surfaces with increases resulting from higher wind speeds. Because the investigators were able to measure the mass load of FO to surfaces as a result of deposition, they were also able to calculate deposition velocities (V_d) with average V_ds to foliar surfaces ranging from 0.0075-0.047 cm/s during exposure duration and relative humidity/rainout tests. Average V_ds of 0.025, 0.081, 0.26, and 0.73 cm/s during 2, 4, 6, and 10 miles per hour wind speeds, respectively, were also calculated and indicate a logarithmic increase in V_d with increasing wind speed in the wind tunnel conditions. The effect of increased wind speeds is considered to be an example of accelerated mass loading resulting from a shift from diffusional and gravitational based deposition to impaction, which results in much higher mass loadings. An average FO deposition velocity to surrogate receptors (filters) during cumulative dose tests was calculated at 0.027±0.003 cm/s. From previous work conducted with red phosphorus which consists of much smaller particles (MMAD=0.75 μm), V_d values among the two smokes were compared and FO's higher calculated V_ds (by a factor of eight) are attributed to the larger particle sizes of FO aerosols, again lending evidence to support limited deposition of particles 0.1 - 1.0 μm through reduced deposition velocity. Deposition velocity is one of a variety of parameters that may be entered into a dispersion model to predict deposition levels at distances downwind from the source/generator.

In a simple Gaussian model used by Driver et al. (1993), for estimation of FO plume and deposition concentrations, deposition velocities (V_d) determined from wind tunnel studies conducted at the Pacific Northwest Labs with FO (0.06 cm/s) (Cataldo et al., 1989) and brass (0.6 cm/s) (Cataldo et al., 1990), were among the parameters entered into the dissemination model for determination of FO deposition downwind from the source (generator). Previously, Driver et al. (1991), while acknowledging lack of significant FO deposition in the field (Liljegren et al., 1988), provided for a worse-case estimate of FO deposition by multiplying airborne FO dosages modeled by the Real-Time Volume Source dissemination model, by a deposition velocity of 0.1 cm/s . The deposition velocity values of 0.06 and 0.6 cm/s from the Cataldo et al. (1989) studies, and 0.1 cm/s which falls in between, are higher than those referenced by Brubaker et al. (1992) at 0.024 - 0.030 cm/s, determined from another wind tunnel study, and 0.0031 cm/s, calculated theoretically. Due to larger particle-size distributions observed under Pacific Northwest Laboratory's wind tunnel conditions in the 1989 report (resulting from coagulation of aged particles), associated V_d values are likely to be unrepresentatively high and therefore may contribute to overly conservative or inaccurate deposition predictions when used in dissemination models.

Deposition concentrations from the RTVS (Driver et al., 1991), Gaussian (Driver et al, 1993), and TECOM (Nester and Imsand, 1993) models (Table 3) were predicted under a variety of atmospheric and generation (source) conditions, as well as deposition velocities, for distances downwind of the generator, and thus, are presented as ranges. The RTVS model (Driver et al.,

1991) provided deposition estimates based on a continuous release of smoke (1.32 gal/min) for 120 minutes (30 minutes x 4 consecutive releases by one generator) and a surface reflection term was entered to indicate complete surface (ground) reflection, which equals no deposition. However, as described above, a worse-case estimate was provided by multiplication of the aerosol dosage by a deposition velocity of 0.1 cm/s. Peak worst-case deposition levels were estimated based on the likelihood of their occurrence within a 90° arc bounded by the directions northwest and northeast as measured from the smoke generator; they are noted in parens following listed levels. Deposition levels for FO were determined to range between 0 and 80 mg/m^2 at 0.1 (30 at 0.2 km), between 0.6 and 6 mg/m^2 at 1 km (3 at 1.5 km) and between about 0.007 and 0.4 mg/m^2 at 10 km (0.3 at 13 km).

TABLE 3. Model-Predicted Deposition Levels of Fog Oil

MODEL DATA
(mg/m^2)

KILOMETERS

	0.1	1.0	4.0	10	40
RTVS MODEL 1 generator; 120" duration at 1.32 gal/min	0 - 80	0.6 - 6		0.007 - 0.4	
GAUSSIAN MODEL 1 generator; 30" duration at 1.33 gal/min	30 - 800	0.3 - 30	0.02 - 3	0.004 - 1	<0.001 - 0.3
TECOM MODEL 6-12 generators; 1' duration at 2-2.2 gal/min	0 - 14	1 - 4	0.1 - 0.6		

The Gaussian model (Driver et al., 1993) that used deposition velocities of 0.06 cm/s and a seemingly high 0.6 cm/s, and a dissemination duration time of 30 minutes per run (1.33 gal/min), generated much higher values in contrast to those predicted by the RTVS model. Predicted deposition levels decreased from 30-800 mg/m^2 at a downwind distance of about 0.1 km, 0.3-30 at 1.0 km, 0.02-3 at 4 km, 0.004-1 at 10 km, to less than 0.001-0.3 mg/m^2 at 40 km. Driver et al. (1993) site hypothetical deposition onto the surface of a body of water at levels of 6,000-60,000 mg/m^2 from a one hour M3A3 generation as determined by Liss-Suter and Villaume (1978). However, these levels were the result of unrealistic assumption of close to 100 percent fall out or settling of all particles disseminated from aerosolization of 150 L of liquid fog oil.

Levels reported by Nester and Imsand (1993) are more similar to those from the RTVS model but incorporate use of greater than one generator per dissemination run (2.2 gal/min). Peak quantities were predicted downwind for six and twelve generators each, for a duration of one hour at neutral and very stable atmospheric conditions, each, at varying wind speeds for a total of eight scenarios. Predictions for deposition along the centerline of the plume decreased from 0-14 mg/m^2 at 0.1 km, 1-4 mg/m^2 at 1.0 km, to 0.1-0.6 mg/m^2 at 4.0 km.

Although all of these models provided predictions of FO deposition, no empirical data has been presented among the reviewed references to substantiate the actual occurrence of measurable deposition outside of about 25 m from the generator, in the field. For purposes of deposition-level prediction, it is imperative that models incorporate input values, such as deposition velocities, that are representative of the true physical characteristics (particle size) of FO aerosols as produced in the field.

4.2.2 Persistence

Physical and chemical characteristics and meteorological conditions determine the persistence of FO droplets in the atmosphere, which has previously been described above in terms of particle size (settling velocity, deposition velocity), density, particle to vapor ratio, and vapor pressure. FO is reported to act as a passive tracer in the atmosphere following turbulent air flow, versus settling (Liljegren et al., 1989), and detectable aerosol concentrations have been measured at out to 4 km at Meadowbrook under stable meteorological conditions (Maloney et al., 1992), and may travel as far as 6.5 km (Brubaker et al., 1992). The theoretical evaporation of a 1.5 μm FO aerosol droplet over time and at various temperatures is discussed in section 2.2, Volatile organic Compounds, but has not been empirically validated for a plume of airborne FO of characteristic particle-size distribution.

In the event of FO deposition to foliar and/or soil surfaces close to the generator in field studies (Liljegren, et al., 1988; Policastro et al., 1989a), or as determined from wind tunnel studies (Cataldo et al., 1989), FO residues are not expected to persist. Cataldo et al. (1989) investigated residence time of FO deposits on foliar, soil, and surrogate (filter) surfaces when mass loading tests produced unexpectedly high variability, indicating losses of FO. FO residues deposited on 47-mm glass fiber filters allowed to evaporate by desiccation and by air, experienced an approximate 14 percent loss of FO mass by each method, due to volatilization during a drying period of 65 days after deposition.

Depuration (loss) rates from environmental surfaces were greater. Investigation of the persistence of FO deposits to Ponderosa pine plants over 9 days following their exposure to FO smoke for 4 hours, resulted in a depuration (loss) curve with two components: a) a rapid loss of FO with a half-time of 1.7 days, and b) an extended half-time that projects beyond the 9-day test duration. Rapid volatilization from the pine is considered to be a result of the relatively large foliar surface area compared to the glass fiber filter. After 4 days, an 80 percent reduction in dose was calculated, however this does not include any losses of dose due to absorbance of FO

constituents into the plant (making them unavailable for extraction); therefore, percent reduction in dose may be higher due to a combination of evaporation and absorbance. Foliar absorption of FO residues was indicated as the origin of transferred, root-retained FO contaminants affecting a reduction in biomass from successive harvests of originally exposed tall fescue plants during residual effects plant tests (Cataldo et al.,1989).

Volatility of FO residues from soils was determined with tests of two soil types over 42 days: 1) Maxey Flats, a silty-clay soil from which FO was lost in two phases, and 2) Burbank, a sandy-loam soil from which FO volatilized monophasically. Loss from Maxey Flats occurred rapidly in the first phase with a half-life of approximately 20 days; the second half-life projected to approximately 500 days. The lower rate of volatilization of FO from Burbank soil was calculated to have an overall half-life of approximately 60 days. Differences in rates of loss are explained by the differing physical characteristics of the soil types where the silty-clay soil has high surface sorption which allowed for an initial increase in volatilization, and the sandy-loam has a more porous consistency with lower total surface area, which resulted in higher downward leaching thereby reducing the initial rate of loss. The FO constituents are expected to biodegrade and lessen in time, specifically undergoing chemical, photochemical, and microbial degradation depending on their location in soils and on other surfaces (Cataldo et al., 1989; Driver et al., 1993). These processes, as well as low deposition velocities, are explained as the reason Brubaker et al. (1992), in an effort to determine contamination of sediment, soil and vegetation with FO residues in areas of heavy use at the Combat Maneuver Training Center, Hohenfels, Germany, were unable to detect FO in any of the samples.

Field or simulated field disseminations of FO smoke aerosols over water bodies resulting in measurement of deposition velocities, deposition levels, and subsequent persistence of contaminants were not available among the literature. Driver et al. (1993) report a hypothetical water-surface deposition level range of 6,000 to 60,000 mg/m^2 for a one hour run of one M3A3 generator. These hefty values are up to 75 times the highest terrestrial level (800 mg/m^2 at 0.1 km in Table 3) determined by dissemination models (previously described as likely overestimating deposition levels) with similar source inputs as those used for determination of the hypothetical levels. Upon closer examination of the original source of the hypothetical water-deposition levels (Liss-Suter and Villaume, 1978), the considerable values appear to be the result of unsophisticated calculations that do not consider settling or deposition velocities, and assume 100 percent precipitation onto surfaces, of 150 liters of completely vaporized FO within an average distance of 1 km downwind of the generator.

Driver et al. (1993) present a review of data on the behavior of "old" fog oil and surrogate oils (such as No. 2 fuel oil and diesel fuel) in water and sediments as determined in laboratory studies which did *not* include aerosol dissemination/deposition. "Old" fog oil is described as slightly mobile in aqueous environments due to the portion of dissolved organic components of (water-soluble fraction) composed mostly of aromatics but which also include a portion of saturated hydrocarbon components. A fractionation or partitioning process occurs with oils in an aqueous environment; the majority of "old" FO constituents are water-insoluble and the insoluble

fraction deposits in the sediment and/or aromatics and insolubles deposit with the aromatics lessening over time due to their solubility. No. 2 fuel oil demonstrated volatility under a constant wind speed with evaporation from petri dishes where components with higher vapor pressures evaporated more readily than those with lower vapor pressures. However, in a study where oil-water dispersions were added to tanks containing sediment and marine water with suspended particulate matter, evaporation as a loss of hydrocarbons was negligible. Oil will also associate with suspended organic matter, sorbing in an inverse proportion to the degree of aqueous solubility; settling of particulate matter adds to the deposition of oil to sediments.

Persistence of FO in natural waters will depend on factors such as the mode of deposition into a body of water, amount of oil, water temperature, sunlight, mixing energy, presence of organic matter and the degree of biotransformation (Driver et al., 1993). In the case of the above No. 2 fuel oil test, biodegradation of hydrocarbons in the sediments began immediately with microbial activity responsible for degradation of the saturated hydrocarbons more readily than the aromatics. A residue of originally present hydrocarbons persisted for at least a year after the end of the experiment. Mitigation efforts during testing and training should substantially protect water bodies from exposure to FO aerosols.

Table 4. Subject Summary By Selected Reference

SUBJECT	Liljegren et al. 1988	Liljegren et al. 1989	Maloney et al. 1992	Policastro et al. 1989 (Annual Report)	DeVaull et al. 1989 (analytical methods)	Policastro et al. 1989 (Models)	Policastro and Dunn 1985	Driver et al. 1991	Driver et al. 1993	Cataldo et al. 1989	Nester and Imsand 1993	Brubaker et al. 1992
Particle Size (PS) (μm); refer to definitions for explanation	average mass median aerodynamic diameter: .74; average GSD: 3.51	average geometric mass mean diameter: .82; average GSD: 1.60	see Liljegren et al. 1989	see previous 3 blocks for PS info covered in this report	mass mean aerodynamic diameter ~0.8; no GSD reported	not reported (review of Liljegren et al., 1988, 1989)	mass median diameters approximately 4.9 and 3.9 depending on generator; no GSD reported	referenced mass median diameters of between about 1 and 2; no GSD reported	determined a theoretical "typical" size: mass median aerodynamic diameter of 1.4 and GSD of 1.65	average mass median aerodynamic diameter: 2.33; average GSD: 1.67	not reported	referenced several particle size distributions and reported particle diameters to be an average of approximately 1
Origin of Aerosol Concentrations	field; Dugway Proving Ground	field; Camp Atterbury	field; Meadowbrook	field	lab scale generator	model data versus field data from DPG, and Camp Atterbury	field	model	model	wind tunnel	equation/ model	historical records
Generator	M3A3E3	M3A4	M3A4	M3A3E3 M3A4	lab scale generator	see Liljegren et al., 1988, 1989	XM49, M3A3, MARS, "Smokey Bear"	modeled XM56 output	modeled XM56 output	lab scale generator	SG18-02	M3A3
Number Generators	1	1	1	1	1	1	1	1	1	1	varies	2

SUBJECT	Liljegren et al. 1988	Liljegren et al. 1989	Maloney et al. 1992	Policastro et al. 1989 (Annual Report)	DeVaull et al. 1989 (analytical methods)	Policastro et al. 1989 (Models)	Policastro and Dunn 1985	Driver et al. 1991	Driver et al. 1993	Cataldo et al. 1989	Nester and Imsand 1993	Brubaker et al. 1992
Release Rate (gal/min)	average= .523	average= .628	average= .762	see Liljegren et al., 1988, '89; Maloney et al., 1992	.000022 to .000053 (oil consumption rate)	see Liljegren et al., 1988, 1989	not reported	1.32	1.33	.000264 to .001584 (oil consumption rate)	1-3	.8
Duration	average= 1 hour	average= 52 min.	10-45 min. per trial	see Liljegren et al., 1988, 1989; Maloney et al., 1992	not reported	see Liljegren et al., 1988, 1989	~5-10 min.	30 min. (x 4 = 120 min. for deposition calculations)	30 min.	2-8 hours (for toxicity testing)	30 min. -1 hr.	15-30 min.
Atmospheric Stability Classes	near neutral to very unstable	near neutral to very unstable	unstable and stable	see Liljegren et al., 1988, 1989; Maloney et al., 1992	not reported	see Liljegren et al., 1988, 1989	extremely unstable to moderately stable	neutral to extreme-ly unstable	extremely unstable to moderately stable	not reported	neutral and stable conditions	not reported
Terrain	simple: uniformly flat	simple: uniformly flat with upwind terrain markedly different	complex: mountain/ valley	see Liljegren et al., 1988, 1989; Maloney et al., 1992	not reported	see Liljegren et al., 1988, 1989	"non-flat"	complex with flat areas suitable for testing	varies	not reported	rolling and rising from sea level to 300 ft	complex: mountain/ valley

SUBJECT	Liljegren et al. 1988	Liljegren et al. 1989	Maloney et al. 1992	Policastro et al. 1989 (Annual Report)	DeVaull et al. 1989 (analytical methods)	Policastro et al. 1989 (Models)	Policastro and Dunn 1985	Driver et al. 1991	Driver et al. 1993	Cataldo et al. 1989	Nester and Imsand 1993	Brubaker et al. 1992
FO	old	old	old	old	old	old	old	old	old	old	old	new
Origin of Deposition Data	field collection; theoretical calculation	theoretical	not reported	field collection; theoretical calculation	not reported	see Liljegren et al., 1988, '89	not reported	model	model	wind tunnel	model	sampled previously heavily used field sites; theoretical calculation
FO Analysis	gross chemical composition	gross chemical composition	gross chemical composition	gross chemical composition	gross chemical composition; qualitative molecular weight distribution	see Liljegren et al., 1988, 1989	analysis of principal chemical element only	citations of previous-ly conducted analyses	citations of previously conducted analyses	total FO measurement	citations of previously conducted analyses	gross chemical composition
Aerosol/ Vapor Ratio	99:1 (field & lab)	99:1 (field & lab)	99:1 (field & lab)	99:1 (field & lab)	99:1 (field & lab)	see Liljegren et al., 1988, 1989	not reported	not reported	not reported; theoretical evaporation over 1 hr. reported as 30% reduc-	not reported	not reported	99:1 as cited from Liljegren et al., 1988, 1989

SUBJECT	Liljegren et al. 1988	Liljegren et al. 1989	Maloney et al. 1992	Policastro et al. 1989 (Annual Report)	DeVaull et al. 1989 (analytical methods)	Policastro et al. 1989 (Models)	Policastro and Dunn 1985	Driver et al. 1991	Driver et al. 1993	Cataldo et al. 1989	Nester and Imsand 1993	Brubaker et al. 1992
Aerosol/ Vapor Ratio Continued									tion in FO-aerosol mass at 40°C			
Ecological Toxicology (Terrestrial and Aquatic)	not reported	not reported	not reported	not reported	not reported	not reported	not reported	cited terrestrial FO studies (plant, soil, earthworm, microbes)	cited terrestrial FO studies (plant, soil, earthworm, microbes) and aquatic studies (as conducted with No. 2 Fuel Oil & FO)	terrestrial FO studies (plant, soil, earthworm, microbes)	cited terrestrial FO studies (plant, earthworm, microbes)	cited terrestrial FO studies

SUBJECT	Liljegren et al. 1988	Liljegren et al. 1989	Maloney et al. 1992	Policastro et al. 1989 (Annual Report)	DeVaull et al. 1989 (analytical methods)	Policastro et al. 1989 (Models)	Policastro and Dunn 1985	Driver et al. 1991	Driver et al. 1993	Cataldo et al. 1989	Nester and Imsand 1993	Brubaker et al. 1992
Fate Pathways	not reported	not reported	not reported	not reported	not reported	not reported	not reported	mobility and dissolution in natural waters, photolysis; fate in soil	in soil, on plants; discussion of bioaccumulation, photolysis, biotransformation, dissolution, sedimentation from citations	in soil, on plants	not reported	in soil, on plants
Notable report features	Well-defined empirical (experimental/field) data including FO aerosol concentra-tions and deposition amounts as a function of	Well-defined empirical (experimental/field) data including FO aerosol concentra-tions and deposition amounts as a function of	Well-defined empirical (experimental/field) data including FO aerosol concentra-tions and deposition amounts as a	Synopsis of the empirical (experimental/field) data from a series of studies (see Liljegren et al., 1988, 1989, and Maloney et	Determination and development of sampling, analysis, and quantification methods for identification of	Evaluation of four Gaussian puff dispersion models using seven data sets collected from trials with fog oil	Evaluation of previously generated field data from product trials of smokes, which results in authors' determination that	Contains both a literature review of available data on FO, FO-aerosols and graphite flakes and	Contains a literature review of previously conducted research on FO and FO aerosols	Contains a series of laboratory tests conducted in an enclosed circulating wind	Review of data generated from past investigations of ecological impacts of fog	Presents data from collection and analysis of field samples from heavily used areas of the site, tested for FO and CS contamina-

SUBJECT	Liljegren et al. 1988	Liljegren et al. 1989	Maloney et al. 1992	Policastro et al. 1989	DeVaull et al. 1989	Policastro et al. 1989 (Models)	Policastro and Dunn 1985	Driver et al. 1991	Driver et al. 1993	Cataldo et al. 1989	Nester and Imsand 1993	Brubaker et al. 1992
Notable report features, continued	source definition (i.e. mass release rate, particle-size distribution), travel distance, meteorological conditions and terrain	source definition (i.e. mass release rate, particle-size distribution), travel distance, meteorological conditions and terrain	function of source definition (i.e. mass release rate, particle-size distribution), travel distance, meteorological conditions and terrain	al., 1992)	the total mass fractions and aerosol concentrations of fog oil (FO) smoke downwind of a source. Additional focus is on gross chemical composition and aerosol size distribution (particle size) of FO aerosols originating from both lab and field generators		the reported data are lacking in quality and reliability for a variety of described reasons	their toxicity and fate, as well as downwind concentrations and surface deposition levels of the materials derived from a dispersion model; no original research presented	and similar oil materials, their health and environmental effects as well as model-predicted downwind aerosol and deposition levels	tunnel, which investigated the terrestrial toxicity and fate of characterized FO aerosols at high concentrations, under varying simulated meteorological conditions	oil and graphite flakes on air, aquatic and terrestrial media, as well as human health; dispersion models are employed to estimate aerosol and deposition levels for use of multiple generators	tion, as well as cited information on nature and effects of the materials to the environment and human health

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GLOSSARY

aspiration: the act of drawing in by suction.

(atmospheric) stability categories (ASC): turbulent diffusing types defined by a set of meteorological conditions. These ASCs may then be used to describe test cases which are further defined by addition of source characteristics (ex. release rate, height of plume release) and other characteristics (ex. deposition velocity).

- ASCs include:
  - A= extremely unstable
  - B= moderately unstable
  - C= slightly unstable
  - D= neutral
  - E= slightly stable
  - F= moderately stable

Meteorological Conditions Defining Pasquill Turbulence Types [from Driver et al. (1993)]

				Nighttime Conditions	
Surface Wind	Daytime Insolation			Thin Overcast or	≤ 3/8
Speed, m/s	Strong	Moderate	Slight	>4/8 Low Cloud	Cloud
<2	A	A-B	B		
2	A-B	B	C	E	F
4	B	B-C	C	D	E
6	C	C-D	D	D	D
>6	C	D	D	D	D

complex terrain/complex meteorology: (ex. Meadowbrook field study) the elevation of the terrain varies significantly (> 100 m); such as when dispersion tests are carried out over mountainous terrain

-measurements must be performed at a number of locations in order to adequately characterize the three-dimensional wind field

convection: the transfer of heat or other atmospheric properties by massive motion directed upward; vertical heat flux

deposition (diffusion) velocity: at a given level of turbulence, the maximum rate of deposition of aerosol particles

- there are two principal ways that small aerosol particles can be removed from an aerosol. The particles can collide with other particles and grow into ones large enough to be removed by gravity or aerodynamic forces (impaction, centrifugal, etc.), or they can migrate to surfaces, stick to those surfaces, and thus be removed. The migration process is known as diffusion.

diffusion: Brownian motion of very small particles sufficient to enhance the probability of hitting object while traveling past it on a non-intercepting streamline

electrostatic attraction: charges on particles/vegetation or substrate

friction velocity: a measure of the shear stress at the surface; a parameter used to define the meteorology of the boundary layer

geometric standard deviation: standard deviation of a log-normal distribution

geometric mean diameter: mean diameter for a log-normal distribution

impaction: when particles with sufficient inertia strike an object placed in the path of the airstream carrying the particles as the airstream changes path direction due to that object

insolation: solar radiation that has been received

interception: particle follows a gas streamline that happens to come within one particle radius of object - hits and is captured

isokinetic: particle paths following air streamlines

-anisokinetic: the deviation of particle paths from the streamlines of the air

log-normal (Ln) distribution: distribution of values X for which Y=Ln(X) is normally distributed (bell curve)

mass median diameter (MMD): the particle size at which 50% of the cumulative mass occurs

mass median aerodynamic diameter (MMAD): the aerodynamic particle size at which 50% of the cumulative mass occurs

- aerodynamic diameter: diameter of a unit density sphere (density = 1 g/cm^3) having the same aerodynamic properties as the particle in question (this means that particles of any shape or density will have the same aerodynamic diameter if their settling velocity is the same); includes inertial characteristics of airborne particles; accounts for effects on particle transport caused by shape of individual particles

- theoretically, FO particles, as spherical liquid droplets, have aerodynamic sizes that equal their physical diameter multiplied by the square root of the specific gravity of FO (~0.92)

- the MMAD is about 4% smaller than the MMD for FO due to aerodynamic consideration of the particles

- the MMDs for FO are greater than the count median diameters (physical median diameter; CMD) due to the log-normal distribution

mean wind: speed and direction of the wind (velocity)

old/new fog oil:

- a 1986 requirement that carcinogens or potential carcinogens be absent from fog oil has necessitated that the oil be treated to remove these compounds by severe hydrotreatment (carbon-carbon double bonds (including aromatic bonds) are hydrogenated and heterocyclics undergo ring opening) or solvent refinement (polynuclear aromatic hydrocarbons (PAHs) and other aromatic compounds including heterocyclics, are removed by solvent extraction); fog oil produced before this requirement is referred to as "old" fog oil, and may contain large numbers of aromatic compounds, while that produced afterward is called "new" fog oil, and should contain only saturated hydrocarbons (aliphatic and cycloaliphatic)

- definitive analysis of the components of "new" fog oil are reported in the Environmental Impact Statement for the Chemical School relocation to Fort Leonard Wood

planetary atmospheric boundary layer: the lowest layer of the atmosphere; meteorology associated with this layer is concerned with the interaction between airflow in the lowest layers of the atmosphere with the planetary surface itself and the associated transfer of energy

PM-10: particulate matter (including FO particles) with a mean aerodynamic diameter of less than or equal to 10 micrometers

PM-2.5: particulate matter (including FO particles) with a mean aerodynamic diameter of less than or equal to 2.5 micrometers

roughness height: roughness = vegetation or other elements such as buildings; if the roughness elements are distributed homogeneously over a site, it is adequate to characterize them in terms of a "roughness height", or height above the ground surface where the wind speed vanishes

(terminal) settling velocity: gravitational settling affect from gravity; the velocity in the direction of gravity

simple terrain/simple meteorology (Dugway field test) = uniformly flat in all directions to the horizon and covered with low, homogeneous roughness elements on the order of 1 cm in height

-measurements may be performed at a single location to adequately characterize the micrometeorology of the entire site

simple terrain/complex meteorology (Atterbury field test) = as terrain or vegetation deviate from above, the micrometeorology becomes more complex:

- if the actual test site remains essentially flat but the upwind terrain is markedly different in elevation or vegetation; an increased roughness height is insufficient to fully characterize the effect of the terrain on the meteorology. The influence of the terrain on the power spectra and statistics of the wind velocity fluctuations can be observed. Yet the microscale meteorology does not vary significantly over the test site.

-measurements may be performed at a single location to adequately characterize the micrometeorology of the entire site

stochastic: a type of dispersion model that employs the convective velocity scale and mixing height as input parameters to account for the rise in the centerline of a smoke plume due to convection (Gaussian models don't account for this rise and therefore may overestimate downwind concentrations most severely after about 250 m in unstable atmospheric conditions)

surface reflection: term used as input into dispersion models to describe percent deposition of material: a range from 0 % (total deposition) to 100 % (no deposition) surface reflection is usually available for entry into calculations that estimate aerosol concentrations and deposition as a function of downwind distance.

transect: specified distances from the source and perpendicular to the prevailing wind containing evenly spaced samplers

volatile organic compound (VOC): The definition of a VOC is any organic compound of carbon (excluding carbon monoxide, carbon dioxide, carbonic acid, metallic carbides or carbonates, and ammonium carbonate) which participates in atmospheric photochemical reactions. A list of further exclusions due to negligible atmospheric photochemical reactivity, is listed in 40 CFR Ch. 51.100 (July 1 1994 edition) under the definition of a VOC.